ADМИXTURES OF NON-WOOD LIGNOCELLULOSIC MATERIALS IN COMBINATION WITH WOOD PARTICLES IN COMPOSITE MATERIALS

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Abstract:
Due to increasing shortage of wood raw materials traditionally used for production of wood based composite materials, there is a growing call for the use of various raw materials from agricultural production. The lignocellulosic residues are plants with rich content of cellulose in their wooden parts or in the fibers and they are used for the production of engineered sheet composites. The big problem is that there is a shortage of these non-wood lignocellulosic materials for the production of composites as well. Therefore, there exist an effort and willingness to utilize non-wood lignocellulosic materials as the substitute of wood at least as admixtures in combination with wood. Various authors indicate the possibilities of the application of non-wood lignocellulosic materials in the particleboard or fiberboard manufacture. The paper deals with the proposal of modification of the particleboard core layer with ratios of hemp shives and cereal straw, the most important alternatives of lignocellulosic materials suitable for the particleboard production in Central European conditions.

Key words: particleboard; hemp; cereal straw; core layer; properties.
INTRODUCTION

Global shortage of wood intended for wood processing industry is continuously deepening, and thus the orientation of research and practice on the search options to find the appropriate partial substituent of wood is needed.

The idea of the possible use of hemp shives and cereal straw as potential substituents of wooden particles portions in particleboard is based not only on the reduced availability of real wood raw material, but also on its continuous prices increase. In the case of a sufficient quantity of hemp shives and cereal straw and in the case of their reasonable prices (which could be at least a little below than wood raw material in corresponding quality), these raw materials should be applicable in the particleboard core layer while retaining the particleboard properties.

NON-WOOD LIGNOCELLULOSIC COMPOSITES

Large amounts of lignocellulosic residues suitable for composites production (about 2.4 billion tons) are produced every year after the end of the agricultural season of the various agricultural species. These residues are either burned down or left over in the ground without any further utilization. In most cases, such residues cause also various disposal problems to the farmers. The agricultural residues are a part of the annual biomass production, and mainly include flax, hemp, bagasse, straws of wheat, barley, oats, rye, rice straw, sugar cane, the stalks of maize, cotton, tobacco, bamboo and the husks or shells of walnuts, almonds etc.

The lignocellulosic residues are excellent raw materials for production of wide range of composites following various chemical or mechanical methods (Kozlowski et al. 2004). In general, the economic feasibility of such uses is influenced by the cost of production, the availability of raw material, and the physical and chemical properties of the lignocellulosic fibers.

Also the growing demand for wood-based panels and the increasing price of wood particles has led to continuous efforts to find new resources as an alternative to wood. The use of other renewable resources such as agricultural residues in the production of composites (i.e. particleboard and fiberboard) has recently been reconsidered as attractive both from the economic and environmental point of view. Value-added lignocellulosic composites made from industrial residues can be considered as an alternative solution to this problem.

Composites based on lignocellulosic particles or fibers can be divided into the following groups (Irle and Barbu 2010):

- Conventional panel-type composites like particleboard, fiberboard, insulating boards etc.;
- Lignocellulosic-mineral composites, which are based on inorganic binders (i.e. cement);
- Natural fibers reinforced polymers;
- Non-woven textile-type composites.

Non-wood lignocellulosic material is an organic residue which consists of mainly cellulose, lignin and hemicelluloses. These structural elements are produced by plants to form the cell walls, leaves, stems, stalks, and woody portions of the plant (Idi and Mohamad 2011).

The literature on wood-lignocellulosic plants composites highlights steady interest for the design of new structures and technologies, towards products for special applications with higher physical and mechanical properties at relatively low prices (Kymäläinen and Sjöberg 2008).

Experimental studies have revealed particular aspects related to the structural composition of lignocellulosic materials, such as:

- The ratio between the different composing elements;
- Their compatibility;
- Types and characteristics of the used resins.

Various technologies have been developed for designing and processing of non-wood lignocellulosic composites by pressing, extrusion, airflow forming, dry, half-dry and wet processes, including thermal, chemical, thermo-chemical, thermo-chemo-mechanical treatments etc. Researchers have undertaken to determine the manufacturing parameters and the physical and mechanical properties of the composites and to compare them with the standard, plywood, particleboard, oriented strand board (OSB), medium density fiberboard (MDF), hardboard, and softboard made from single wood raw material (Irle et al. 2012).

Limitations of the lignocellulosic residues than can be used in the production of composites are as follows:

- The primary drawback of the use of agro-fibers is the lower processing temperature permissible due to possible of lignocellulosic degradation and/or volatile emissions that could affect composite properties;
- The second drawback is the high moisture absorption of the natural fibers. Moisture absorption can result in swelling of the fibers, and concerns about the dimensional stability of the agro-fiber composites cannot be ignored (Sanadi et al. 1996);
- The major limitations of using agricultural by-products are the lack of an established collection, storage and handling systems that would prevent the degradation of the lignocellulosics when stored for a considerable period (Reddy and Yang 2005).

Real available quantities of hemp shives or cereal straw are not sufficient to produce a meaningful pure board, and therefore it was considered the idea of a mixture of particles and shives (or cereal straw) in a certain ratio in the particleboard core construction. From visual reasons, it was not considered with an addition of shives to the particleboard surface layers.

Modification of the core layer of particleboard in the theoretical level can be made from two reasons:

1. Improvement of the performance properties of finished particleboard through the addition of a new component to their core layer;
2. Replacement of the part of particle mass in the particleboard core layer by other material.

In the first case, by adding of a new ingredient to the particleboard core layer, it is followed the synergistic effect of multiplying the board properties requested for their specific use, e.g. increased resistance of boards against moisture content, water, fire, biotic or abiotic agents, increased strength properties, etc. In the latter case, the aim is the raw materials cost savings and the effort to incorporate "cheaper" material into the particleboard. Principles of the particleboard core layer modifications are basically the same (Réh and Vrtielka 2012).

Several authors have tried to incorporate the different raw material than wood particles into the particleboard construction. Detailed study of literature sources on the use of non-wood lignocellulosic plants for composite materials production was compiled by Youngquist et al. (1994). The possibility to replace wood particles by hemp shives in the production of particleboard and the possibility to produce lightweight composite boards is discussed by Schopper et al. (2009). They reduced the density of three-layer particleboard using hemp shives from 650 kg.m⁻³ to 400 kg.m⁻³. Pecenka et al. (2010) state the growing global demand for high quality boards made from flax or hemp as alternative raw material in industries such as automotive, construction and furniture.

The most important lignocellulosic residues in Central Europe are flax (Linum usitatissimum L.) and cereal straw (Triticum aestivum L.).

MATERIALS AND METHODS

Similar or same technological parameters were used for the production of modified particleboard, as are applied in practice for standard particleboard production:

- Dimensions of the boards prepared in the laboratory: width 300 mm, length 400 mm, thickness 16 and 18 mm
- Number of boards for each alternative: 5 pieces
- Particles reserve attributable to the production of each series of five boards: 30%
- Board density: 620 – 630 kg.m⁻³
- Board moisture content after conditioning: 7%
- Ratio of surface to core particles: 2:3
- Moisture content of surface particles: 4.5%
- Moisture content of core particles, shives and cereal straw: 2%
- Adhesive amount to surface particles: 10%
- Adhesive amount to core particles, shives and cereal straw: 6%
- Adhesive dry matter: 65%
- Paraffin emulsion amount to the surface particles: 0.47%
- Paraffin emulsion amount to the core particles and shives: 0.27%
- Paraffin emulsion dry matter: 37%
- Hardener amount to the surface particles: 0.8%
- Hardener amount to the core particles, shives and cereal straw: 3.5%
- Hardener dry matter: 58%
- Pressing temperature: 220 °C
- Compression factor 8 s.mm⁻¹.

Laboratory production procedure of standard particleboard, modified particleboard, and determination and evaluation of their physical and mechanical properties:
- Preparation and analysis of input materials
- Drying of shives, cereal straw, wood surface and core particles
- Sorting of surface and core particles
- Preparation adhesive mixture and auxiliary chemicals
- Adhesive mixture application and auxiliary chemicals
- Mat forming
- Prepressing
- Pressing
- Cooling and conditioning
- Manufacture of test specimens
- Measurement and evaluation of selected physical properties (moisture content, thickness swelling after 2 hours, thickness swelling after 24 hours) and mechanical properties (bending strength, modulus of elasticity in bending, tensile strength perpendicular to face) according to the appropriate EN.

Mat forming was done manually; the following ratios of wood particles, shives, and cereal straw were used for the particleboard core layer:

- Ratio no. 1: 0% of shives (or cereal straw) – 100% particles
- Ratio no. 2: 5% of shives (or cereal straw) – 95% particles
- Ratio no. 3: 10% of shives (or cereal straw) – 90% particles
- Ratio no. 4: 15% of shives (or cereal straw) – 85% particles
- Ratio no. 5: 20% of shives (or cereal straw) – 80% particles
- Ratio no. 6: 25% of shives (or cereal straw) – 75% particles
- Ratio no. 7: 30% of shives (or cereal straw) – 70% particles

RESULTS AND DISCUSSION

The average density of pressed modified particleboard was 625±10 g.m⁻³. The ratio of shives or cereal straw in the particleboard core layer affected the density in almost insignificant extent.

According to the testing methods provided in the standard EN 322:1995 the average percentage of particleboard moisture content is ranging from 8 to 12% (www.europeanpanels.eu, www.ewp.asn.au). All the percentage ratios of shives or cereal straw particleboard as well as the unmodified particleboard meet the prescribed moisture content. Moisture content of shives or cereal straw particleboard was not significantly higher than unmodified particleboard moisture content. It was not confirmed the statistically significant rate correlation between the moisture content and the ratio of shives or cereal straw addition in particleboard at the significance level \( \alpha = 0.05\% \) based on regression analysis.

Results of bending strength, modulus of elasticity and tensile strength perpendicular to face for shives or cereal straw particleboard as well as for unmodified particleboard samples are shown in Table 1, results of swelling after 2 and 24 hours are shown in Table 2.

Based on results of tables 1 and 2 it may be concluded that mechanical tests did not confirm the negative influence of the wood substituents except too high concentration of cereal straw in particleboard.

**Table 1**

<table>
<thead>
<tr>
<th>Ratio shives or cereal straw to wood particles (%)</th>
<th>hemp (MPa)</th>
<th>cereal straw (MPa)</th>
<th>hemp (MPa)</th>
<th>cereal straw (MPa)</th>
<th>hemp (MPa)</th>
<th>cereal straw (MPa)</th>
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</thead>
<tbody>
<tr>
<td>Bending strength</td>
<td>14.4</td>
<td>3 600</td>
<td>2 800</td>
<td>0.65</td>
<td>0.41</td>
<td></td>
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<tr>
<td>Modulus of elasticity in bending</td>
<td>3 700</td>
<td>2 750</td>
<td>2 850</td>
<td>0.62</td>
<td>0.38</td>
<td></td>
</tr>
<tr>
<td>Tensile strength perpendicular to face</td>
<td>3 300</td>
<td>2 700</td>
<td>2 800</td>
<td>0.61</td>
<td>0.32</td>
<td></td>
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<tr>
<td></td>
<td>3 400</td>
<td>2 750</td>
<td>2 800</td>
<td>0.60</td>
<td>0.24</td>
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<td></td>
<td>3 450</td>
<td>2 800</td>
<td>2 800</td>
<td>0.58</td>
<td>0.23</td>
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<tr>
<td></td>
<td>3 100</td>
<td>2 200</td>
<td>2 800</td>
<td>0.52</td>
<td>0.18</td>
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</tbody>
</table>
Table 2

Swelling after 2 and 24 hours for shives or cereal straw particleboard and for unmodified particleboard samples

<table>
<thead>
<tr>
<th>Ratio shives or cereal straw to wood particles (%)</th>
<th>Hemp (MPa)</th>
<th>Cereal straw (MPa)</th>
<th>Hemp (MPa)</th>
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<tr>
<td>Swelling after 2 hours</td>
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<tr>
<td>0 – 100</td>
<td>11.5</td>
<td>34.0</td>
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<td>5 – 95</td>
<td>15.1</td>
<td>12.3</td>
<td>39.7</td>
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<td>10 – 90</td>
<td>15.8</td>
<td>14.2</td>
<td>39.2</td>
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<tr>
<td>15 – 85</td>
<td>20.0</td>
<td>15.8</td>
<td>39.0</td>
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<tr>
<td>20 – 80</td>
<td>23.1</td>
<td>16.4</td>
<td>38.6</td>
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<tr>
<td>25 – 75</td>
<td>23.8</td>
<td>17.3</td>
<td>38.5</td>
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<tr>
<td>30 – 70</td>
<td>24.2</td>
<td>18.6</td>
<td>37.3</td>
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<td>Swelling after 24 hours</td>
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According to the testing methods provided in the standard EN 310:2011 typical value in three-point bending strength for standard particleboard is 11.0 - 15.0 MPa depending on particleboard type (www.europeanpanels.eu, www.ewp.asn.au). The minimum value of bending strength prescribed is met for almost all ratios of shives and cereal straw except too high concentration of cereal straw in particleboard. The addition of shives and cereal straw affect bending strength of particleboard positively, particleboard with the addition of shives and cereal straw in core layer has shown even higher bending strength than unmodified particleboard except too high concentration of cereal straw in particleboard. Bending strength slightly decreases with the ratio of 20% and more which may be due to a higher ratio of shives or cereal straw that are more flexible than wood particles. It was not confirmed statistically significant degree of correlation between bending strength and ratio of shives or cereal straw addition in particleboard to the significance level $\alpha = 0.05\%$ based on regression analysis.

Typical value for modulus of elasticity in bending is 1 600 - 2 600 MPa depending on particleboard type (www.europeanpanels.eu, www.ewp.asn.au). This value is met for all ratios of shives and cereal straw. Based on regression analysis it was confirmed the statistically significant degree of correlation between modulus of elasticity in bending and shives or cereal straw ratio addition in particleboard on the significance level $\alpha = 0.05\%$.

According to the testing methods provided in the standard EN 319:1995 typical value of tensile strength perpendicular to face for particleboard with thickness of 16mm is specified at the level of 0.35 - 0.45 MPa depending on particleboard type (www.europeanpanels.eu, www.ewp.asn.au). This minimum value was complied with all shives ratios. Tensile strength perpendicular to face was decreasing with the increasing percentage of shives in particleboard. This phenomenon is probably due to the different shives geometry compared with wood chips. For cereal straw, the minimum value complied with the ratio of 5 only which excludes the use of particleboard with higher ratio of cereal straw in some applications. This condition can be caused not only due to other cereal straw geometry compared with wood chips, but also due to the chemical composition of cereal straw. Based on regression analysis it was confirmed the statistically significant degree of correlation between tensile strength perpendicular to face and shives ratio addition in particleboard on the significance level $\alpha = 0.05\%$.

According to the testing methods provided in the standard EN 317:1999 allowed particleboard swelling after 2 hours is up to 8% (www.europeanpanels.eu, www.ewp.asn.au). All swelling results are above this limit for all monitored ratios of shives and cereal straw, and even for unmodified particleboard. We assume that this result could be caused by amount of hydrophobic agent added during the manufacturing process or by fractional composition of surface and core particles in particleboard. It applies for swelling after 2 hours, that swelling values are increasing with an increasing percentage of shives and cereal straw, because of the different chemical composition of shives and cereal straw compared with wood particles because shives and cereal straw are able to absorb more water compared with wood particles. Based on regression analysis it was confirmed the statistically significant degree of correlation between swelling after 2 hours and shives and cereal straw ration addition in particleboard on the significance level $\alpha = 0.05\%$.

Swelling after 24 hours had reversed course compared to swelling after 2 hours. While swelling after 2 hours rose with an increasing proportion of shives, swelling after 24 hours has shown decreasing tendency. It can probably be caused due to the presence of shives that after a certain period started to take less water than particles and thus slowed down the swelling course. Based on regression analysis it was confirmed the statistically significant degree of correlation between swelling after 24 hours and shives ratio in particleboard on the significance level $\alpha = 0.05\%$. 
CONCLUSIONS
The experiment was aimed to the analysis of the modification impact of core layer in the production of particleboard with the addition of shives and cereal straw ratio on selected physical and mechanical properties. Conditions for the production of particleboard and for the implementation experiment were set up to resemble to the conditions of normal particleboard production. From the analyses performed it can be concluded that there is a statistical dependence between the modifications of particleboard core layer by cereal straw and its physical and mechanical properties. It was also confirmed that non-wood lignocellulosic raw materials may not be used for energy purposes only, but they may have their real use in the production of wood based composites.

Modification of the particleboard core layer with ratios of hemp shives and cereal straw, the most important alternatives of lignocellulosic materials suitable for the particleboard production in Central European conditions, is possible. It is possible to produce particleboard with the substitution of wood particles in the core layer with maximum 20 – 30% of hemp shives and with maximum 10 – 15% of cereal straw without the negative change of particleboard properties or without change of the technical and technological production parameters.

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